

PCT

WORLD INTELLECTUAL PROP.  
International B

INTERNATIONAL APPLICATION PUBLISHED UNDER

WO 9606885A1

(51) International Patent Classification 6 : C08K 5/521, C08L 75/04	A1	(11) International Publication Number: <b>WO 96/06885</b> (43) International Publication Date: 7 March 1996 (07.03.96)
(21) International Application Number: PCT/EP95/03397 (22) International Filing Date: 29 August 1995 (29.08.95) (30) Priority Data: 94202474.6                30 August 1994 (30.08.94)        EP (34) Countries for which the regional or international application was filed:                NL et al. (71) Applicant (for all designated States except US): AKZO NOBEL N.V. [NL/NL]; Velperweg 76, NL-6824 BM Arnhem (NL). (72) Inventors; and (75) Inventors/Applicants (for US only): BLUNDELL, Cefn [GB/NL]; Paulus Potterlaan 10, NL-1213 EX Hilver- sum (NL). WUESTENENK, Johan, Antonie [NL/NL]; Haitinkweg 6, NL-7021 KB Zelhem (NL). (74) Agent: SCHALKWIJK, Pieter, Cornelis; Akzo Nobel N.V., Patent Dept. (Dept. APTA), P.O. Box 9300, NL-6800 SB Arnhem (NL).	(81) Designated States: JP, US, European patent (AT, BE, CH, DE, DK, ES, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE). Published With international search report. Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.	
(54) Title: FOG REDUCTION IN POLYURETHANE FOAM USING PHOSPHATE ESTERS (57) Abstract <p>A process for imparting flame retardancy to a polyurethane foam comprising the step of incorporating in said polyurethane foam 0.5-50.0 php of at least one flame-retardant phosphate ester is disclosed. Also disclosed is a flame-retardant polyurethane foam composition which comprises a polyurethane foam and 0.5-50.0 php of at least one flame-retardant phosphate ester, as well as the use of a flame-retardant phosphate ester to impart flame retardancy to a polyurethane foam. An example of such a phosphate ester is resorcinol bis(diphenyl phosphate). These flame retardants have the advantage that they reduce fogging caused by evaporation of volatile components from polyurethane foam compositions.</p>		

**FOR THE PURPOSES OF INFORMATION ONLY**

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

AT	Austria	GB	United Kingdom	MR	Mauritania
AU	Australia	GE	Georgia	MW	Malawi
BB	Barbados	GN	Guinea	NE	Niger
BE	Belgium	GR	Greece	NL	Netherlands
BF	Burkina Faso	HU	Hungary	NO	Norway
BG	Bulgaria	IE	Ireland	NZ	New Zealand
BJ	Benin	IT	Italy	PL	Poland
BR	Brazil	JP	Japan	PT	Portugal
BY	Belarus	KE	Kenya	RO	Romania
CA	Canada	KG	Kyrgyzstan	RU	Russian Federation
CF	Central African Republic	KP	Democratic People's Republic of Korea	SD	Sudan
CG	Congo	KR	Republic of Korea	SE	Sweden
CH	Switzerland	KZ	Kazakhstan	SI	Slovenia
CI	Côte d'Ivoire	LI	Liechtenstein	SK	Slovakia
CM	Cameroon	LK	Sri Lanka	SN	Senegal
CN	China	LU	Luxembourg	TD	Chad
CS	Czechoslovakia	LV	Larvia	TG	Togo
CZ	Czech Republic	MC	Monaco	TJ	Tajikistan
DE	Germany	MD	Republic of Moldova	TT	Trinidad and Tobago
DK	Denmark	MG	Madagascar	UA	Ukraine
ES	Spain	ML	Mali	US	United States of America
FI	Finland	MN	Mongolia	UZ	Uzbekistan
FR	France			VN	Viet Nam
GA	Gabon				

## FOG REDUCTION IN POLYURETHANE FOAM USING PHOSPHATE ESTERS

## FIELD OF THE INVENTION

5

The present invention relates to anti-fogging flame retardants useful in polyurethane foams and to a process for imparting flame retardancy to a polyurethane foam by incorporating such anti-fogging flame retardants into the polyurethane foam.

10

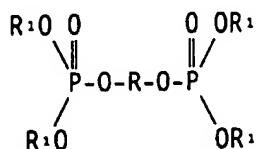
## BACKGROUND OF THE INVENTION

15

U.S. Patent No. 4,746,682 to J. Green relates to a polyurethane flame retardant containing a polybrominated diphenyl oxide and an alkylated triaryl phosphate ester.

20

Certain disclosures exist in the prior art in regard to organic diphosphates of the general class



25

30

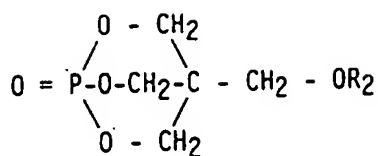
where R can be substituted or unsubstituted alkylene, arylene, alkylene arylene, or other divalent connecting radical (e.g., diphenylene sulfone -SO-, -S-, -SO<sub>2</sub>-), and the like and R<sub>1</sub> can be substituted or un-substituted aryl or alkyl. Examples of some disclosures include the following: U.S. Patent Nos. 3,492,373; 3,869,526 and 4,203,888; British Patent No. 2,061,949; and Japanese Patent Publication Nos. 57/174,331; 71/22,584; 74/40,342 and 82/55,947.

Also relating to this class of compounds are copending U.S. Patent Application Serial No. 374,717, filed July 3, 1989, entitled "Novel Aromatic Bisphosphates" which covers compounds where R<sub>1</sub> is aryl and R is -C<sub>6</sub>H<sub>4</sub>SO<sub>2</sub>H<sub>4</sub>- and U.S. Patent Application Serial No. 374,716, filed July 3, 1989, also entitled "Novel Aromatic Bisphosphates" which covers compounds where R<sub>1</sub> is also aryl and R is a neopentyl group or a halogenated neopentyl group.

Most of these flame retardants suffer from the drawback that when incorporated in polurethane compositions, they are too volatile or contain volatile impurities which will be released when the polyurethane is heated. This is becoming an increasingly significant problem in the automobile industry because the new aerodynamic designs employ, for example, a flatter windscreen which increases the surface area of sun penetration into the auto on warm, sunny days. This often leads to internal temperatures above 100°C and thus evolution of volatile products from polyurethane parts in the auto. These volatile products produce a thin film in the auto which is particularly noticeable on the windscreen. This phenomena has been termed, "fogging."

Automakers are beginning to demand polyurethanes which exhibit significantly less fogging in order to eliminate this problem. Accordingly, there is a need for flame retardant compositions which provide adequate flame retardancy in polyurethane foam compositions and reduce the amount of fogging which occurs upon heating of these compositions. This and other objects of the present invention will become apparent from the summary and detailed description which follow.

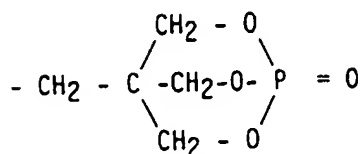




5 (III)

wherein  $R_2$  is selected from hydrogen,  $C_6$ - $C_{20}$  aryl,  $C_6$ - $C_{20}$  substituted aryl,  $-\text{P}(\text{O})(\text{OR}_1)_2$  wherein  $R_1$  is as defined above, and the group:

10



15

In a second aspect, the present invention relates to polyurethane foam compositions which comprise 0.5-50.0 php of at least one flame retardant phosphate ester represented by the formula I. Finally, the present invention also relates to the use of phosphate esters of the formula I as flame retardants in polyurethane foam.

Compounds of the formulas I-III and methods for making them are known. For example, the product Fyrolflex® RDP (ex. Akzo Nobel Chemicals) is commercially available for use as a flame retardant plasticizer in thermoplastic resins such as nylons, polyesters, polycarbonates and blends thereof. It is known to demonstrate excellent thermal stability and compatibility with thermoplastic resins. Further, the compound of the formula I where  $n=1$ , which is known as phosphoric acid 1,3-phenylene tetraphenyl ester or resorcinol bis(diphenyl phosphate), has CAS Registry Number 57583-54-7. Thus, methods for making this compound and its higher oligomers ( $n=2$ -10) are known in the art.

25  
30

Preferred are compounds of the formula I. More preferred are compounds of the formula I wherein R<sub>1</sub> is a C<sub>6</sub>-C<sub>20</sub> aryl group, optionally halogenated. The preferred compounds for use in the present invention are resorcinol bis(diphenyl phosphate), neopentyl glycol bis(diphenyl phosphate), propylene glycol bis(diphenyl phosphate), bisphenol A bis(diphenyl phosphate), and mixtures thereof. Resorcinol bis(diphenyl phosphate) is the most preferred flame retardant for use in the present invention. In the present invention, 0.5 - 50.0 php of the flame retardant of the formulas I-III is employed in a polyurethane foam to impart flame retardancy thereto. More preferably, 3-25 php of the flame retardant is employed and, most preferably, 4-15 php of the flame retardant is used. These amounts of the flame retardants of the present invention not only impart some flame retardancy to the polyurethane but they also reduce the amount of fogging as compared to the same polyurethane without flame retardant and/or including other flame retardant compounds.

The present invention is applicable to all polyurethanes. However, it is preferred that the present invention be applied to polyurethanes derived from aromatic isocyanates such as toluene diisocyanate and diphenylmethane diisocyanate. The second component of the polyurethanes is a polyol, preferably selected from polyetherpolyols and polyesterpolyols, with polyurethanes derived from polyetherpolyols being preferred due to their processing ease. In the most preferred embodiment, the present invention is applied to flexible polyurethane foams since this type of polyurethane foam is typically used in applications where fogging problems must be eliminated.

The flame retardants of the present invention are generally incorporated in the polyurethane by mixing them in during the production of the polyurethane. The present flame retardants are thereby homogeneously mixed into the foam matrix. Methods for polyurethane production are well known to those of skill in the art.

Other additives can be incorporated into the polyurethane foam along with the flame retardants of the present invention. More particularly, a catalyst is typically employed in the production of flexible polyurethane foams. Such catalysts are generally a mixture of one or more tertiary amines with an organo-tin catalyst.

5 Further, chain extenders and/or cross-linking agents may also be employed in the polyurethane foam production process in order to tailor the properties of the foam to the application for which it will be used. For example, using chain extenders the modulus and the glass  
10 transition temperature can be raised and using cross-linking agents, the foam can be made harder and stronger.

Of course, a foaming agent is required in the production of polyurethane foams, such as, for example, water.

15 Another component in the polyurethane foam is the surface active agent which is generally employed to improve the mixability of the reaction mixture and to stabilize the gas bubbles thereby providing more uniform cells in the foam. Typical surface active agents include  
20 polysilicones and the type and amount of polysilicone is chosen on the basis of the type of foam that is desired.

The polyurethane foam may also include both organic and inorganic coloring agents which both provide the desired foam color and offer  
25 some protection against ultraviolet light. Carbon black is a typical coloring agent employed in such foams. The foam may also include one or more fillers in order to make the foam heavier and/or stronger. Fibrous and inorganic mineral fillers may be used in this application.

30 Finally, it is possible to combine the flame retardants of the formulas I-III of the present invention with one or more conventional flame retardants used in polyurethane foams whereby the increase in fogging caused by the conventional flame retardants can be offset by



the flame retardants of the present invention. Typical flame retardants which can be used are halogenated diphenyl oxides, haloalkyl phosphates and phosphonate flame retardants such as those which are commercially available for use in polyurethane foams.

5 The invention will be further illustrated by the following examples.

#### Test Methods

10 Fogging was measured according to DIN 75201 using the gravimetric method. The limiting oxygen index (LOI) was measured according to ISO 4589. The automotive flame test was carried out according to MVSS 302. The rise time is the time at which the foam ceases to rise during its production.

15 The air flow test measures the ease with which air flows through the cellular polyurethane structure. In this manner, the openness of the cells can be determined. The test samples were 51 mm x 51 mm x 25 mm. The sample is placed in an opening and the amount of air stream required to produce a pressure drop of 12.70 mm water over the sample  
20 is measured.

#### Polyurethane Foam Production Method

25 The polyol, flame retardant, water, amine catalysts and the stabilizer were mixed, with stirring, in a first beaker. In a separate beaker, the toluene diisocyanate (TDI) was weighed out. The box wherein the foam will be made was prewarmed and the organo-tin catalyst was put into a syringe. The first beaker was stirred at 1800 revolutions per  
30 minute for a period of 5 seconds and then the organo-tin catalyst was dosed thereto while stirring was continued. After a total of 21 seconds of stirring, the TDI was added to the mixture. Stirring was then continued for an additional 9 seconds and the still fluid mixture

was quickly put into the box. Then the rise time was measured. Once the foam ceased to rise, it was subjected to an additional 10 minutes at 110°C to ensure that the reaction was completed.

#### 5 Example 1 and Comparative Examples A-B

10 A polyurethane foam formulation was prepared from 100 parts of Lupranol® 2030, 3.5 php water, 12 php of the flame retardant mentioned in Table 1, 0.15 php Dabco® 33LV, 0.05 php Dabco® BL11, 1.2 php Tegostab® B3640, 0.4 php Dabco® T10 and 46.57 php of toluene diisocyanate (110 index). The fogging, rise time, limiting oxygen index (LOI), specific gravity, and direct air flow for these materials were measured and the results are presented in Table 1. Further, these compositions were subjected to the automotive flame test and it was found that the composition in accordance with the present invention (Example 1) was self-extinguishing.

Table 1

20 Example	Flame Retardant	Fogging (mg)	rise time	LOI (%O <sub>2</sub> )	specific gravity	automotive flame test	air flow direct
A	none	0.9-1.0	97	18.8	----	B/RB	----
B	Phosflex® TPP	1.2-1.9	72	21.8	33.7	SE	42.5
1	RDP	0.4-0.6	150	21.2	33.0	SE	17.5

25 B/RB = Burned  
SE = Self-extinguishing  
LOI = Limiting Oxygen Index

30 These results clearly show that the flame retardant of the present invention leads to less fog than the prior art flame retardant and even produces a reduction in fogging when compared to the control which contained no flame retardant.

Examples 2-5 and Comparative Examples 2a-5a and 4b

In these examples, polyurethane foams were prepared from 4 different polyols as are given in Table 2. The foam formulations are the same as for Example 1 except that different polyols were employed. Examples 2a-5a are control examples wherein the fogging of polyurethane foams containing no flame retardant were measured and the results appear in Table 2 under the heading, "control." Examples 2-5 incorporate 12 php of resorcinol bis(diphenyl phosphate).  $\Delta$ Fog is the difference between the amount of fogging from the control example and the amount of fogging for the same composition including 12 php of the flame retardant of the present invention. The results appear in Table 2.

Example	Polyol in Polyurethane Foam	Table 2 Fogging in mg		$\Delta$ Fog
		Control	RDP (12 php)	
2-2a	Caradol® 1080	1.7	1.1	-0.6
3-3a	Caradol® 36-3	1.0	0.5	-0.5
4-4a	Caradol® 48-2	2.0	1.5	-0.5
4b	Caradol® 48-2	2.0		+0.2-0.3
5-5a	Lupranol® 2030	0.9	0.5	-0.4

RDP = Resorcinol bis(diphenylphosphate)

Example 4b was performed using Armguard® V6, a commercially available flame retardant for polyurethane foams.

The results in Table 2 demonstrate that the flame retardants of the present invention actually produce a net reduction in fogging when added to a polyurethane foam whereas one would expect a net increase in fogging when a flame retardant is added.

Examples 6-8

The polyurethane foam formulation of Example 1, containing as polyol, Caradol® 48-2, was tested with three different amounts of resorcinol bis(diphenyl phosphate) [RDP] flame retardant. The results are given in Table 3.

Table 3

	Example	RDP (php)	$\Delta$ FOG in mg
10	6	12	-0.5
	7	6	-0.5
	8	2	0

The results in Table 3 demonstrate that different amounts of the flame retardant of the present invention produce the unexpected effect that there is either no net increase in fogging due to the addition of the flame retardant, or, at higher concentrations, a net decrease in fogging can be realized.

Example 9 and Comparative Examples C-D

In these examples, the formulation of Example 1 was tested using flame retardants at 12 php levels in a polyurethane foam derived from Lupranol® 2030. The results are given in Table 4.

Table 4

Example	Flame Retardant	$\Delta$ FOG in mg
C	Antiblaze® 100	1.4
D	DEG-CEP	0.6-1.1
5 9	NPG-TPP	0.1-0.3

Antiblaze® 100 is a commercially available polyurethane flame retardant. DEG-CEP is a diethylene glycol based chloroethyl phosphate. 10 NPG-TPP is neopentyl glycol bis(diphenyl phosphate).

From the results in Table 4 one can see the expected significant increase in fogging which is normally a side-effect of incorporating conventional flame retardants into polyurethane foams. In comparison, 15 the flame retardant in accordance with the present invention demonstrate a reduced fogging when compared to Antiblaze® 100 and DEG-CEP. These results clearly demonstrate that the fogging reduction achieved by the flame retardants of the present invention is significant and unexpected.

20

#### Examples 10-11

In these examples, the formulation of Example 1 was employed except 25 that Lupranol® 2030 was replaced by the polyol Caradol® SC-5602, the amount of flame retardant in accordance with the present invention was reduced to 8 php and the flame retardants propylene glycol bis(diphenyl phosphate) (PG-TPP) and bisphenol A bis(diphenyl phosphate) (BPA-TPP) were employed in the tests. The results are 30 given in Table 5.

12

Table 5

Example	Flame Retardant	$\Delta$ FOG in mg
10	PG-TPP	0.28-0.38
11	BPA-TPP	0-0.2

5

From the results in Table 5 one can see that other flame retardants in accordance with the present invention also give an unexpectedly good reduction in fogging.

10

Example 12

Example 10 was repeated except that 6 php of PG-TPP was employed. The results are given in Table 6.

15

Table 6

Example	Flame Retardant	$\Delta$ FOG in mg
12	PG-TPP	0.16-0.35

20

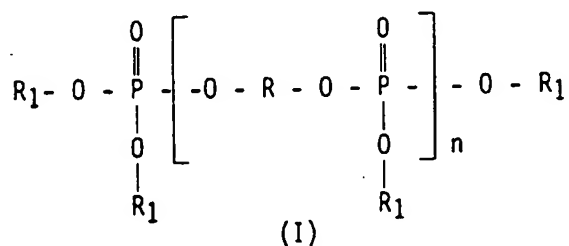
The foregoing examples were presented for the purpose of illustration and description only and are not to be construed as limiting the invention in any way. The scope of the invention is to be determined from the claims appended hereto.

25

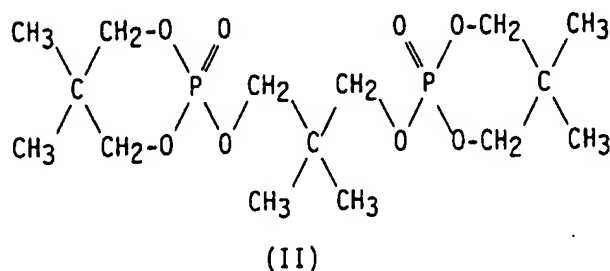
30

What is Claimed is:

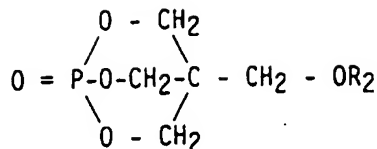
1. A process for imparting flame retardancy to a polyurethane foam comprising the step of incorporating in said polyurethane foam 0.5-50.0 php of at least one flame retardant phosphate ester represented by the formulas I-III:



wherein  $n=1-10$ ; R is selected from linear or branched  $C_2-C_{20}$  alkyl, and  $C_6-C_{20}$  aryl, optionally halogenated; and each  $R_1$  is independently selected from linear or branched  $C_1-C_{10}$  alkyl,  $C_6-C_{20}$  aryl, and  $C_7-C_{30}$  aralkyl, the aryl and aralkyl groups being optionally halogenated;

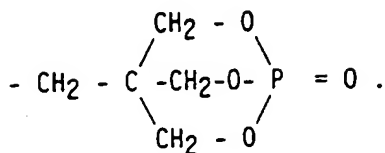


and,



(III)

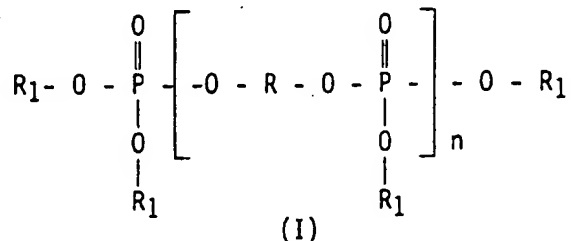
wherein  $R_2$  is selected from hydrogen,  $C_6$ - $C_{20}$  aryl,  $C_6$ - $C_{20}$  substituted aryl,  $-P(O)(OR_1)_2$  wherein  $R_1$  is as defined above, and the group:



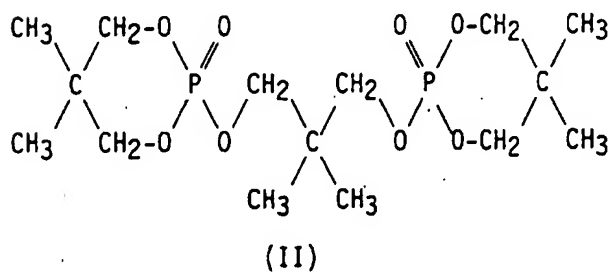
2. A process in accordance with claim 1 wherein 3-25 php of said flame retardant of the formulas I-III is employed.
3. A process in accordance with any one of claims 1-2 wherein in said process a second flame retardant, other than a flame retardant of the formulas I-III, is incorporated into said polyurethane foam.
4. A process in accordance with claim 3 wherein said second flame retardant is selected from halogenated diphenyl oxides, haloalkyl phosphates and phosphonate flame retardants.
5. A process in accordance with any one of claims 1-4 wherein said flame retardant of the formulas I-III is resorcinol bis(diphenyl phosphate).



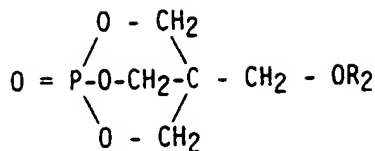
6. A flame retardant polyurethane foam composition which comprises a polyurethane foam and 0.5-50.0 php of at least one flame retardant phosphate ester represented by the formulas I-III:



wherein  $n=1-10$ ; R is selected from linear or branched  $\text{C}_2\text{-C}_{20}$  alkyl, and  $\text{C}_6\text{-C}_{20}$  aryl, optionally halogenated; and each  $\text{R}_1$  is independently selected from linear or branched  $\text{C}_1\text{-C}_{10}$  alkyl,  $\text{C}_6\text{-C}_{20}$  aryl, and  $\text{C}_7\text{-C}_{30}$  aralkyl, the aryl and aralkyl groups being optionally halogenated;



and,

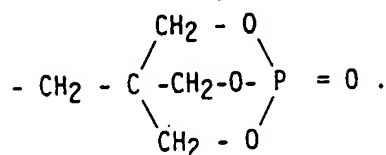


5

(III)

wherein  $R_2$  is selected from hydrogen,  $C_6$ - $C_{20}$  aryl,  $C_6$ - $C_{20}$  substituted aryl,  $-P(O)(OR_1)_2$  wherein  $R_1$  is as defined above, and the group:

10



15

7. A composition in accordance with claim 6 which comprises 3-25 php of said flame retardant of the formulas I-III.

20

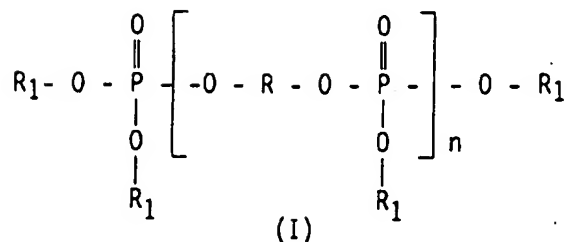
8. A composition in accordance with any one of claims 6-7 which comprises a second flame retardant other than a flame retardant of the formulas I-III.

25

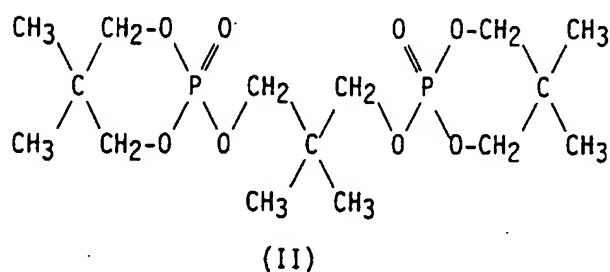
9. A composition in accordance with any one of claims 6-8 wherein said flame retardant of the formulas I-III is resorcinol bis(diphenyl phosphate).

30

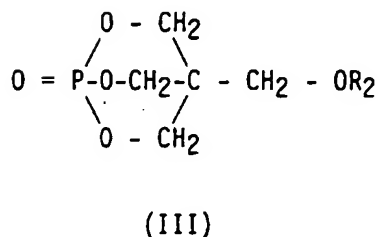
10. Use of a flame retardant phosphate ester represented by the formulas I-III:



wherein  $n=1-10$ ;  $R$  is selected from linear or branched  $C_2-C_{20}$  alkyl, and  $C_6-C_{20}$  aryl, optionally halogenated; and each  $R_1$  is independently selected from linear or branched  $C_1-C_{10}$  alkyl,  $C_6-C_{20}$  aryl, and  $C_7-C_{30}$  aralkyl, the aryl and aralkyl groups being optionally halogenated;

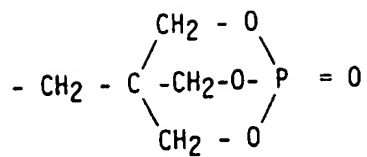


and,



wherein  $R_2$  is selected from hydrogen,  $C_6-C_{20}$  aryl,  $C_6-C_{20}$  substituted aryl,  $-P(O)(OR_1)_2$  wherein  $R_1$  is as defined above, and the group:

18



5

to impart flame retardancy to polyurethane foam.

10

15

20

25

30

## INTERNATIONAL SEARCH REPORT

International Application No  
PCT/EP 95/03397

A. CLASSIFICATION OF SUBJECT MATTER  
IPC 6 C08K5/521 C08L75/04

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 C08K

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
P,X	DATABASE WPI Week 9503 Derwent Publications Ltd., London, GB; AN 95-019429 & JP,A,06 306 277 (DAIHACHI CHEM IND) , 1 November 1994 see abstract ---	1,5,10
Y	FR,A,2 338 283 (MICHIGAN CHEM. CORP.) 12 August 1977 see page 2, line 17 - line 24; claims 1,7; examples ---	1,2,10
Y	US,A,2 952 701 (R.L. MCCONNELL ET AL.) 13 September 1960 see column 2, paragraph 3 --- -/--	1,2,10

☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

## \* Special categories of cited documents :

- "A" document defining the general state of the art which is not considered to be of particular relevance
- "E" earlier document but published on or after the international filing date
- "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- "O" document referring to an oral disclosure, use, exhibition or other means
- "P" document published prior to the international filing date but later than the priority date claimed

- "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
- "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
- "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.
- "&" document member of the same patent family

Date of the actual completion of the international search

8 December 1995

Date of mailing of the international search report

15. 01. 96

Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2  
NL - 2280 HV Rijswijk  
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,  
Fax: (+31-70) 340-3016

Authorized officer

Engel, S

## INTERNATIONAL SEARCH REPORT

Internu Application No

PCT/EP 95/03397

## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	EP,A,0 224 979 (G.F.C. FOAM CORP.) 10 June 1987 see page 19 - page 20; examples XVI,XVII ---	1,2,10
A	US,A,3 412 051 (CH.F. BARANAUCKAS) 19 November 1968 see column 7 - column 8, line 25; table 1 -----	1,2,10

1

This Page is inserted by IFW Indexing and Scanning  
Operations and is not part of the Official Record

## BEST AVAILABLE IMAGES

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images include but are not limited to the items checked:

- ☐ BLACK BORDERS
- ☐ IMAGE CUT OFF AT TOP, BOTTOM OR SIDES
- ☐ FADED TEXT OR DRAWING
- ☐ BLURED OR ILLEGIBLE TEXT OR DRAWING
- ☐ SKEWED/SLANTED IMAGES
- ☐ COLORED OR BLACK AND WHITE PHOTOGRAPHS
- ☐ GRAY SCALE DOCUMENTS
- ☒ LINES OR MARKS ON ORIGINAL DOCUMENT
- ☐ REPERENCE(S) OR EXHIBIT(S) SUBMITTED ARE POOR QUALITY
- ☐ OTHER: \_\_\_\_\_

**IMAGES ARE BEST AVAILABLE COPY.**

**As rescanning documents *will not* correct images problems checked, please do not report the problems to the IFW Image Problem Mailbox**